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ESTIMATION OF RADIOLOGICAL EXPOSURE LEVELS IN A MINING AREA BASED ON ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K ACTIVITY MEASUREMENTS: A CASE STUDY FOR BEYLIKOVA-SIVRIHISAR COMPLEX ORE SITE IN TURKEY

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The study estimated the radiological exposure levels in a mining area for miners in the Beylikova-Sivrihisar (Turkey) complex ore site containing ^{238}U, ^{226}Ra and ^{232}Th. Sixty samples were collected from the study area based on a geologic map. The radionuclide activities were measured using a 78.5% efficient n-type HPGe detector. The measured mean activities were 1871 ± 38 Bq kg^{-1} for ^{238}U, 1749 ± 5 Bq kg^{-1} for ^{226}Ra, 3467 ± 9 Bq kg^{-1} for ^{232}Th and 309 ± 2 Bq kg^{-1} for ^{40}K. From the measured results, the external effective dose was calculated to be max. 3.80 ± 0.03 mSv y^{-1} (mean: 2.04 ± 0.03 mSv y^{-1}) for inside gallery and max. 7.59 ± 0.05 mSv y^{-1} (mean: 4.08 ± 0.05 mSv y^{-1}) for outdoor. Additionally, the external exposure index was calculated to be H_{ex} = 33.5 ± 0.2(mean: 18.2 ± 0.3) and internal exposure index to be H_{in} = 45.9 ± 0.3 (mean: 22.9 ± 0.4). The results indicate that additional radiation protection measures should be taken during the mining process if an opencast mining is implemented in Beylikova complex ore deposit.

INTRODUCTION

Today, it is well known that activity concentrations of radionuclides such as ^{238}U, ^{210}Pb, ^{226}Ra, ^{232}Th and ^{40}K in soil, stream, lake and sea sediments and other environmental samples might contribute to the total radiation dose exposure of humans. In this context, the radioactivity measurements in mining samples can provide valuable information about the radiation exposure levels faced by miners due to naturally occurring sources in the mining region, thus leading to accurate assessments of their health risk. However, the radiation exposure due to naturally occurring radionuclides and their decay products varies greatly depending on the radionuclide concentrations of rocks and soils that form the geological structure of the region[(1)]. In direct measurements of the radionuclides in various samples, the gamma-ray spectrometry with HPGe detector is commonly preferred over other nuclear analytical techniques as it is quite easy and simple from the standpoint of sample preparation and is non-destructive and provides rapid and more accurate results[(2)].

It is an important point to investigate and evaluate radiological exposure levels faced by miners in addition to the radioactive content of the mining site. This is because the workers are exposed not only to radionuclide-bearing dust particles but also to the gamma-ray emissions from those radionuclides during the several mining processes. It is worth noting that mining sites can contain much more uranium, thorium and potassium deposits than anticipated. Therefore, it is imperative that the potential radiological exposure levels faced by workers are determined based on deposit ore samples before a mining operation can be licensed.

The gamma spectrometric radioactivity measurements can provide information on the external and radiation exposure indices. From these factors, the radiological exposure levels can be estimated. The permissible individual dose limits are 1 mSv/y for people and maximum 50 mSv/y in a single year or average 20 mSv/year over for radiation workers, also for mining workers[(19,28)].

It also should be noted that the radon measurements in mining galleries and open side areas are not directly included in this study. That is, the internal exposure index calculated from gamma-radioactivities of ^{226}Ra, ^{232}Th and ^{40}K radionuclides is an indicative factor.

The complex ore mining area, namely, Beylikova-Sivrihisar in Turkey was chosen as the study area because it has a remarkable ^{238}U and ^{232}Th content. This mining area has an apparent reserve of 53 million tons complex ore, which consists of 37.44% fluorite, 31.04% barite, 0.2% thorium oxide and 3.14% rare earth oxides[(3)]. The main ore minerals of the deposit

are fluorite, barite and bastnaesite. Thorium is found in bastnaesite minerals. The average ThO_2 amount is 0.2%, but in some samples, ThO_2 content was reported to exceed 3% in the ore site[3]. The main problem is that radioactive elements such as ^{232}Th and ^{238}U-^{226}Ra and their decay products contained in this deposit may cause serious health problems to miners during mining operations such as during the separation of minerals (fluorite, barite and bastnaesite) and processing of rare earth element (REE) oxides.

Possible radiation dose rates are directly related to the radionuclide activity in ore material, and the mining workers might be exposed externally to the gamma-emitting radionuclides during the mineral processing steps like cracking, grinding, baking, leaching etc. In addition to the risk posed by external radiation, there is also internal irradiation exposure due to normal breathing in mine site. Normal breathing would contain much more radon (^{222}Rn: half-life: 3.8232 (8) d), thoron (^{220}Rn: half-life: 55.8 (3) s) concentration in the inhaled air and their decay products bound to aerosols and dust particles. If the activity levels of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K are known in advance, the probable radiation exposure levels for mining workers can be reduced. However, this requires accurate activity measurements of the radionuclide. This will enable special attention and precautions to be considered at the mine site before mining operations proceed.

The main objective of this study is to estimate the external gamma dose levels and exposure indices to mining workers inside the gallery. However, the activity results measured from the samples collected from the outcropping ore surfaces (mostra) can also provide information about radiological exposure level indices for the case of an open-cast mining operation. A dose rate calculation model is employed by taking into account the miner's working hours and the measured activity concentrations in samples, collected from the study area of a geologic map of Beylikova-Sivrihisar in Northwestern Anatolia, Turkey. In this work, the activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in samples were measured with a high resolution gamma-ray spectrometry.

MATERIALS AND METHOD

Description of study area

Beylikova-Sivrihisar complex ore site is located between Kızılcaören—Karkın—Okçu villages, in the southeast of Eskisehir province, in northwestern Anatolia, Turkey. There is no settlement in this region. The total area is 17.2 km^2, and this area is separated into seven sectors by Eti Maden Mine Corporation (The current license holder) as Devebağırtan, Koca devebağırtan, Yaylabaşı, Küçükhöyüklü, Canavarini, Kocayayla and Köyyeri considering mineralisation characteristics of the area. Historically, complex ore deposit at the site was discovered towards the end of 1959 by the Turkish Establishment of Mining Exploration (MTA) during aerial investigations for uranium exploration studies. This area is the biggest REE oxide deposit of Turkey and also one of the largest thorium deposits in the world with an apparent reserve of 380,000 tonnes thorium oxide[4,5]. In Turkey, this deposit is called REE resources in which REE oxides can be recovered economically and thorium can also be attained as a by-product. The geological map of the study area is shown in Figure 1.

Sampling procedure

Sixty samples were collected from seven different locations under a supervising geology expert of Eti Maden mine corporation on 08 November 2018. The geographic positions of the sampling locations were defined by GPS and recorded. B6 and B7 samples are surface soil samples taken from outside the complex ore site. Ore samples between B1–1 and B1–24 were taken from outside a gallery in the Devebağırtan sector, and ore samples between B1–25 and B1–35 were taken from inside of this gallery. The samples collected from complex ore site were packed in proper plastic and cotton bags, labeled and transferred to the laboratory.

Sample preparation procedure

The samples were separately dried, crushed and milled to obtain fine grains. The powdered samples were homogenised using a ring grinder, made of tungsten. Then they were sieved (<75 μm particle size) with Retsch brand AS-300 sieve, filled into a 500 mL Marinelli beaker container and weighed with a balance (AND brand) that provided an accuracy of ±0.1 mg. All containers were sealed with silicon glue, labeled with the required info and stored for at least 2–3 months to achieve radioactive secular equilibrium in radium-radon decay chain.

Radioactivity measurements

The radioactivity contents of the collected samples were measured with an n-type HpGe detector (ORTEC GMX70P4-S) with a 78.5% relative efficiency and a P/C ratio of 74:1 at 1332.5 keV of ^{60}Co, and its energy resolution (FWHM) is 0.8 keV at 122.1 keV of ^{57}Co and 2.08 keV at 1332.5 keV of ^{60}Co. The detector is already installed in the centre of a 10 cm lead shield lining with Sn and Cu to minimise X-rays and natural background radiation in the laboratory. The gamma spectrometry system connected to a 16 K digital analyser (Ortec DSPEC Jr.2.0) and the detector is biased an detector interface module(Ortec DIM)[7]. The spectrum is acquired and

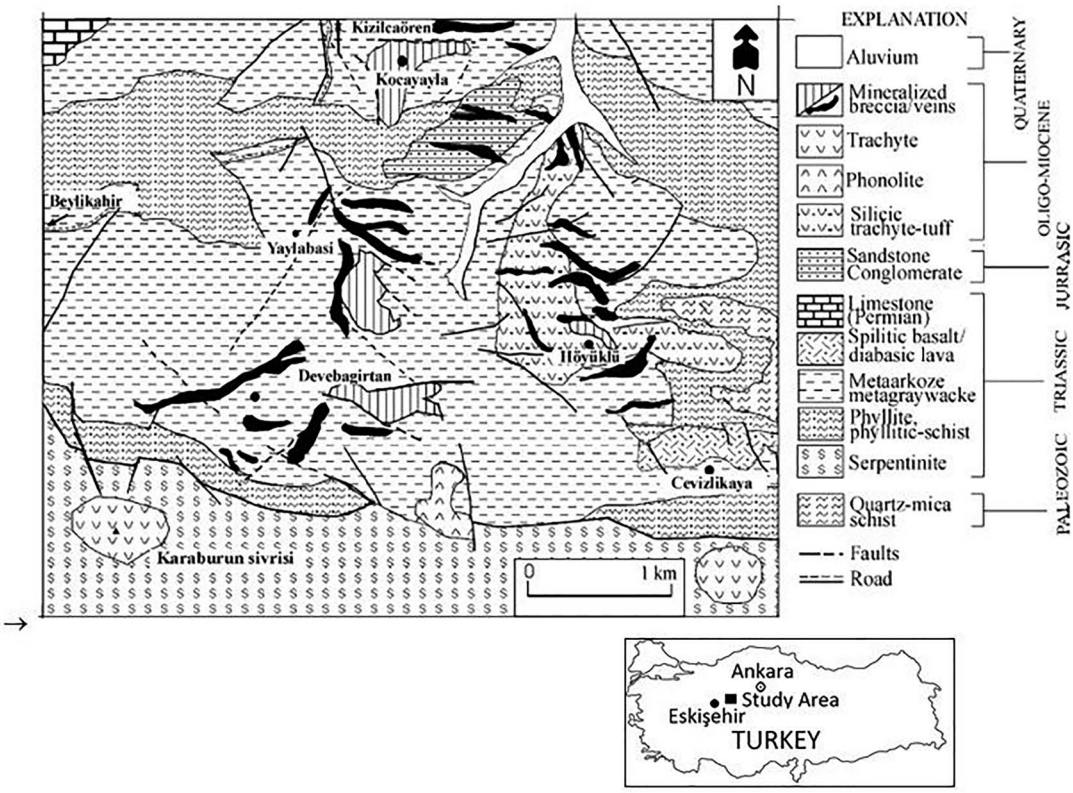

Figure 1: geologic map of study area (modified from Gültekin et al.[6]).

analysed by the Gamma Vision gamma spectroscopy software.

The quality control of the calibrated spectrometer was performed prior to present research work using IAEA-certified standard samples containing uranium, thorium and potassium such as RGU-1, RGTh-1 and RGK-1 and certified CUP-1, CUP-2, BL-2, BL-5 and DH-1 as well as OKA-2 (obtained from CANMET NRC, Canada) that contained REE element oxides. These were measured within the accuracy of less than ±5% for checking U, Th and K amounts.

^{238}U itself is the main radionuclide of the uranium decay series, but it emits very weak gamma rays that are difficult to measure in 49.55 keV (0.0697(26) %) and 113.5 keV (0.0174(47) %) energy[8]. Therefore, the activity of ^{238}U is assumed to be in equilibrium with product nuclide ^{234m}Pa (1.159 minutes)[9,10]. Therefore, after correcting for proper background contribution, it is a reasonable method for the activity determination of ^{238}U, the 1001 keV (0.847(8)%)[8] gamma-ray peak of ^{234m}Pa is the best one since it has also less self-absorption effect.

It is a fact that ^{214}Pb and ^{214}Bi do not quickly achieve equilibrium with ^{226}Ra because the precursor product ^{222}Rn (3.82 days) may require more than 30 days to reach equilibrium with ^{226}Ra, and this needs to be with ^{214}Pb and ^{214}Bi in a closed system. ^{226}Ra itself emits main 186.2 keV (3.55%) gamma ray, but this peak needs to correct for the spectral contribution of 185.7 keV(57.0(3)%) of ^{235}U isotope that contains in natural uranium at least 0.71% wt. ^{235}U[11]. However, the ^{214}Pb decay product has also the most intense peaks at 295.2 keV (18.414(36)%) and 351.9 keV (35.60(7)%) gamma emissions. Similarly, the ^{214}Bi decay product has the most intense peaks at 609.3 keV (45.49(19)%) and 1120.2 keV (14.91(3)%)[8]. Therefore, in a geological sample, ^{226}Ra activity was easily determined by taking the arithmetic mean of either its own peak (186.2 keV) and/or its decay products ^{214}Pb (295.2 keV and 351.9 keV) peaks and ^{214}Bi (609.3 and 1120.2 keV) peaks. However, the spectral interference correction to the peak area of 186.2 keV(^{226}Ra) should be made for determining a more accurate activity value of 186.2 keV gamma rays because the spectral

contribution of 185.7 keV (^{235}U) overlaps with the analytical peak of 186.2 keV (3.55%). Thus, ^{226}Ra activity does not require the radioactive equilibrium condition with subsequent decay products such as ^{214}Pb and ^{214}Bi [11].

In this work, these measurements were corrected for spectral interference using the methodology described in previous works[11,12]. True coincidence corrections should also be made for 609.3 and 1120.2 keV peaks of ^{214}Bi especially in a close counting geometry where a Marinelli beaker is used [7].

Where radioactive equilibrium exists, ^{232}Th activity can easily be determined from its decay products such as ^{228}Ac and ^{208}Tl. The most intense gamma rays of ^{228}Ac are 338.4 keV (11.4(4)%), 911.1 keV (26.2(8)%) and 968.9 keV (15.9(5)%). The most intense gamma energy of ^{208}Tl is 583.1 keV (85.0(3)%) and 860.5 keV (12.4(1)%) when their emission probabilities are multiplied by its branching ratio of 35.93%. ^{232}Th activity was calculated by taking the arithmetic mean of the related photopeak of the decay products ^{228}Ac (338.4 and 911.1 keV) peaks and ^{208}Tl (583.1 and 860.5 keV) peaks by correcting for true coincidence effects, especially in Marinelli beaker geometry because it is a close detection geometry[7].

Potassium activity is simply measured from photopeak energy of 1460.82 keV (10.55(11)%) of ^{40}K nuclide. In most cases, this peak is assumed to be clean; however, the 1460.8 keV might have interference 1459.13 keV (0.87(5)%) gamma-ray of ^{228}Ac[8]. This spectral interference correction is very important to determine accurate ^{40}K activity if the sample contains also thorium[13]. The spectral contribution of 1459.13 keV (^{228}Ac) overlapping to the analytical peak of 1460.82 keV (^{40}K) can simply be estimated from the measured activities by the following formula:

$$F_{sp} = \left[1 + 0.08246 \times \frac{\mathrm{A}\left(^{228}\mathrm{Ac}\right)}{\mathrm{A}\left(^{40}\mathrm{K}\right)}\right] \quad (1)$$

After analysing the gamma-ray spectra acquired with a calibrated high resolution gamma-ray spectrometer described above, the activity of the radionuclides of interest in the samples was determined by the well-known formula

$$A = \frac{N_p/t_c}{\varepsilon(E) m\, P_\gamma(E)} \cdot \left[F_c \cdot F_{sp} \cdot F_s\right] \quad (2)$$

where A is activity (Bq·kg^{-1}), N_p is the net counts after subtracted background counts, t_c is the live-time (s) for the measurement, ε(E) is the photopeak efficiency at a given energy, m (kg) is sample dry mass, P_γ(E) is the gamma-emission probability at a given energy, taken by Decay Data Evaluation Project (DDEP) of LNHB[8], F_c is the true coincidence correction factor, F_{sp} is spectral interference factors for some peaks and F_s is self-absorption factors for all analytical peaks of interest.

Each sample was counted for at least 2–3 days to provide sufficient counting statistics. In this work, GESPECOR (Ver 4.2) software based on MCNP code simulation was used to calculate true coincidence correction and gamma self-absorption correction factors. In the simulation model, all features were described taking into account all geometric and physical parameters of the detector and material properties[14].

For the estimation of self-absorption effects, it is necessary to know the elemental composition of the samples. To do this, an EDXRF system consisting of a 500 μm Silicon Drift Diode (SDD) detector (FWHM = 132 eV@5.9 keV, 40 nm window) made of Si_3N_4 material and an excitation source with 50 kVp X-ray tube with Ag-anode[15] was used to determine the elemental contents in mining samples collected from Beylikova-Sivrihisar site. The EDXRF method can be applied in many different ways. In this work, it was used by adopting the standard comparison method in which OKA-2 certified reference material (rare earth-thorium ore containing 2.893 ± 0.058 wt.% Th and 218.6 ± 8.2 μg/g U) was chosen because its approximate elemental composition is known. It was obtained from **CCRMP, CanmetMINING(**Canadia). In sample preparation, about 1–2 g sample was mixed with a small amount of cellulose binder and then pressed into a pellet form. From the X-ray spectra acquired with the EDXRF system with SDD, the characteristic X-rays of the elements can easily be analysed by using K_α and K_β line intensities through a special analysis software (XRS-FP Ver. 5.7, provided by Amptek Inc., USA), and then they are compared with those obtained from the standard samples, which are generally with a very close matrix to the sample[16,17]. Those obtaining elemental concentrations are then used in GESPECOR software as input data for each sample, in order to calculate self-absorption coefficients for the relevant analytically used gamma-ray energies in this study.

RESULTS

The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K of the complex ore samples obtained from the region of Beylikova-Sivrihisar are given in Table 1. The range of activity concentrations for ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K varied from 63 ± 23 to 4793 ± 54 Bq kg^{-1} (mean value 1871 ± 38 Bq kg^{-1}),

from 410 ± 1 to 7373 ± 12 Bq kg^{-1} (mean value 3467 ± 9 Bq kg^{-1}), from 52 ± 0.3 to 6030 ± 10 Bq kg^{-1} (mean value 1749 ± 5 Bq kg^{-1}) and from 40 ± 2 to 1149 ± 5 (mean value 309 ± 2 Bq kg^{-1}), respectively. The highest mean ^{238}U and ^{226}Ra activity was observed in B2 location and the highest mean ^{232}Th activity was observed in B5 location.

It was observed that the activity of ^{232}Th is always above the certain world average value of 40 Bq kg^{-1} [18]. ^{232}Th activity is highly independent of the location in this Beylikova-Sivrihisar region. The same findings cannot be referred to ^{238}U, ^{226}Ra and ^{40}K activity. They varied greatly in different locations. Therefore, it can be concluded that ^{232}Th is the more common ore in this area.

B6 and B7 group samples taken from outside of the complex ore site are surface soil samples. Therefore, it is seen that these samples have lower activity values when compared with the samples taken from other sections because of the absence of ore content. Additionally, it is observed that no radioactive equilibrium conditions existed in surface samples from B-6 and B7 group.

In the Devebağırtan sector of the mining site, there is a 70–80 m deep gallery excavated during previous research. Ore samples between B1–1 and B1–24 were taken from outside of this gallery (i.e. open area) and ore samples between B1–25 and B1–35 were taken from inside of this gallery. The radioactive equilibrium condition can be simply quantified by an equilibrium factor as the activity ratio of $\delta = A(^{238}U)/A(^{238}Ra)$. For inside the gallery, the radioactive equilibrium condition is mostly attained, δ = 1.00–1.20 for 11 ore samples coded as B1–25 to B1–35. As expected, some outliers calculated for $^{238}U/^{226}Ra$ radioactive equilibrium $\delta = 2.23$ for B3–1, δ = 1.45 for B3–2 and δ = 1.82 for B4–2 samples. The outliers for radioactive equilibrium $\delta = 0.79$ for B1–6, $\delta = 0.78$ for B1–19 imply that radium leaching might be expected in ore site in long geological times. It appears that the ore samples are in radioactive equilibrium but individual results from near surface and soil samples may show marked deviation from the equilibrium. This implies that the ore deposit is near surface (outcropping/mostra), weathering process might result in disequilibria and dose profile may be different than the one anticipated.

Uniform distribution of naturally occurring nuclides at a height of 1 m from the ground, absorbed dose rate, D (in nGy h^{-1}) due to gamma-ray exposure depending on the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K natural radioisotopes is calculated by the following formula[18]:

$$D = 0.462\,A(^{226}Ra) + 0.604\,A(^{232}Th) + 0.0417\,A(^{40}K) \quad (3)$$

where dose conversion coefficients(in nGy/h per Bq/kg) are 0.462 for ^{238}U, 0.604 for ^{232}Th and 0.0417 for ^{40}K, respectively[25]. A(^{226}Ra), A(^{232}Th) and A(^{40}K) are radium, thorium and potassium activity concentrations (in Bq kg^{-1}), respectively.

For open area (outdoor) and inside the gallery calculations, annual external effective dose rate (in mSv y^{-1}) due to gamma-rays emitted from the radionuclides of ^{226}Ra, ^{232}Th and ^{40}K are calculated by the following formulae given in UNSCEAR (2000) report[18]:

$$ED_{out\_gallery} = D\left(nGy\,h^{-1}\right) \times 2000\,h\,y^{-1} \times 0.7 \times 10^{-6} mSv\,nGy^{-1} \quad (4)$$

$$ED_{inside\_gallery} = D\left(nGy\,h^{-1}\right) \times 1000\,h\,y^{-1} \times 0.7 \times 10^{-6} mSv\,nGy^{-1} \quad (5)$$

This external exposure from gamma emitting radionuclides is a component of the total annual effective dose from all sources of radiation. 0.7 Sv/Gy is the dose conversion factor from absorbed dose in the air to the effective dose. For the outside-the-gallery(outdoor) calculations, miners are assumed to work eight (8) hours a day, five (5) days a week and fifty (50) weeks a year. For the inside gallery calculations, miners are assumed to work four (4) hours a day, five (5) days a week and fifty (50) weeks a year. Due to the variability of mining operation areas, the annual external effective dose rate should be calculated not only for inside the gallery but also for outdoor(open mining area).

The radioactivity levels are generally of ambient background levels and use of terms like 'hazard' is an exaggeration. The H_{ex} and H_{in} values need to be less than unity for insignificant effect on health. However, even if they are ten times higher, they do not pose any serious adverse effects or cause any significant health hazards except that the risk of stochastic effect may increase marginally[26]. Hence, notable International reports[25,19,28] such as UNSCEAR (2008), ICRP 103 (2007) and IAEA GSR Part III (2011) do not use the word 'hazard' for such ambient exposure, except that they specify limits for annual doses or radioactivity levels. The review of Rao[26] suggests external exposure index (H_{ex}) and internal exposure index(H_{in}) instead of the terminology 'health hazard indices'. Additionally, the H_{ex} and H_{in} parameters of unity correspond to 1.5 mGy/y of radiation exposures[27]. For possible occupational exposure of the workers in this mining area, exposure indices are important parameters that can provide valuable information to assess the exposure levels[20]. External exposure index (i.e. external health hazard index) H_{ex} can be

Table 1. ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K activity concentrations, absorbed dose rates, external effective dose for inside the gallery, external effective dose for outside the gallery, internal exposure index and external exposure index.

Sample code	Activity concentrations A (Bq kg^{-1})*				Absorbed dose, D (nGy·h^{-1})	Effective dose for inside gallery	Effective dose for outside gallery	Internal Exposure Index	External Exposure Index
	^{238}U	^{232}Th	^{226}Ra	^{40}K	D (nGy h^{-1})	ED_{in} (mSv y^{-1})	ED_{out} (mSv y^{-1})	H_{in}	H_{ex}
B1–1	1443 ± 40	3723 ± 9	1348 ± 4	116 ± 2	2876 ± 51	2.01 ± 0.04	4.03 ± 0.07	21.7 ± 0.4	18.0 ± 0.3
B1–2	1929 ± 45	4801 ± 11	1787 ± 5	141 ± 2	3731 ± 55	2.61 ± 0.04	5.22 ± 0.08	28.2 ± 0.4	23.4 ± 0.3
B1–3	3278 ± 20	1299 ± 5	3457 ± 6	57 ± 1	2384 ± 42	1.67 ± 0.03	3.34 ± 0.06	23.7 ± 0.4	14.4 ± 0.3
B1–4	1480 ± 19	1427 ± 5	1365 ± 2	145 ± 3	1499 ± 32	1.05 ± 0.02	2.10 ± 0.04	12.9 ± 0.3	9.2 ± 0.2
B1–5	1771 ± 29	4404 ± 8	1587 ± 3	149 ± 1	3399 ± 24	2.38 ± 0.02	4.76 ± 0.03	25.6 ± 0.2	21.3 ± 0.2
B1–6	4793 ± 54	1284 ± 7	6030 ± 10	40 ± 2	3563 ± 135	2.49 ± 0.09	4.99 ± 0.19	37.6 ± 1.4	21.3 ± 0.8
B1–7	2950 ± 38	1197 ± 5	3044 ± 6	43 ± 2	2131 ± 75	1.49 ± 0.05	2.98 ± 0.10	21.1 ± 0.7	12.9 ± 0.5
B1–8	2680 ± 40	2646 ± 7	2645 ± 6	86 ± 2	2824 ± 66	1.98 ± 0.05	3.95 ± 0.09	24.5 ± 0.6	17.4 ± 0.4
B1–9	2516 ± 29	3707 ± 8	2450 ± 5	124 ± 1	3376 ± 29	2.36 ± 0.02	4.73 ± 0.04	27.6 ± 0.2	21.0 ± 0.2
B1–10	2253 ± 14	3520 ± 6	2149 ± 3	110 ± 1	3124 ± 29	2.19 ± 0.02	4.37 ± 0.04	25.2 ± 0.2	19.4 ± 0.2
B1–11	1695 ± 24	3162 ± 7	1560 ± 4	89 ± 1	2634 ± 31	1.84 ± 0.02	3.69 ± 0.04	20.7 ± 0.2	16.4 ± 0.2
B1–12	3444 ± 45	3690 ± 9	2926 ± 6	112 ± 2	3585 ± 65	2.51 ± 0.05	5.02 ± 0.09	30.1 ± 0.5	22.2 ± 0.4
B1–13	2151 ± 29	4829 ± 9	1920 ± 4	150 ± 1	3810 ± 28	2.67 ± 0.02	5.33 ± 0.04	29.1 ± 0.2	23.9 ± 0.2
B1–14	1819 ± 34	4459 ± 10	1684 ± 4	147 ± 2	3477 ± 49	2.43 ± 0.03	4.87 ± 0.07	26.3 ± 0.4	21.8 ± 0.3
B1–15	1940 ± 20	2038 ± 5	1736 ± 3	116 ± 1	2038 ± 19	1.43 ± 0.01	2.85 ± 0.03	17.3 ± 0.2	12.6 ± 0.1
B1–16	3414 ± 44	2061 ± 7	3229 ± 6	107 ± 2	2741 ± 52	1.92 ± 0.04	3.84 ± 0.07	25.4 ± 0.5	16.7 ± 0.3
B1–17	1915 ± 32	4860 ± 10	1701 ± 4	151 ± 1	3728 ± 27	2.61 ± 0.02	5.22 ± 0.04	28.0 ± 0.2	23.4 ± 0.2
B1–18	2276 ± 27	5643 ± 11	2084 ± 4	170 ± 1	4378 ± 28	3.06 ± 0.02	6.13 ± 0.04	33.1 ± 0.2	27.5 ± 0.2
B1–19	2072 ± 23	1750 ± 5	2668 ± 5	57 ± 1	2292 ± 41	1.60 ± 0.03	3.21 ± 0.06	21.2 ± 0.4	14.0 ± 0.2
B1–20	3237 ± 43	2985 ± 8	3132 ± 6	110 ± 2	3255 ± 46	2.28 ± 0.03	4.56 ± 0.06	28.5 ± 0.4	20.0 ± 0.3
B1–21	1749 ± 14	2434 ± 5	1554 ± 5	149 ± 1	2194 ± 17	1.54 ± 0.01	3.07 ± 0.02	17.8 ± 0.1	13.6 ± 0.1
B1–22	2449 ± 35	3066 ± 7	2470 ± 8	102 ± 2	2997 ± 60	2.10 ± 0.04	4.20 ± 0.08	25.2 ± 0.5	18.5 ± 0.4
B1–23	3090 ± 38	1943 ± 5	2992 ± 6	60 ± 2	2558 ± 86	1.79 ± 0.06	3.58 ± 0.12	23.7 ± 0.8	15.6 ± 0.5
B1–24	1960 ± 37	3954 ± 9	1763 ± 5	142 ± 2	3209 ± 47	2.25 ± 0.03	4.49 ± 0.07	24.8 ± 0.4	20.1 ± 0.3
B1–25	1912 ± 49	3943 ± 11	1918 ± 6	134 ± 2	3273 ± 51	2.29 ± 0.04	4.58 ± 0.07	25.6 ± 0.4	20.4 ± 0.3
B1–26	2263 ± 52	3970 ± 11	2123 ± 6	147 ± 2	3385 ± 48	2.37 ± 0.03	4.74 ± 0.07	26.8 ± 0.4	21.1 ± 0.3
B1–27	2151 ± 16	4265 ± 6	2051 ± 3	124 ± 1	3529 ± 29	2.47 ± 0.02	4.94 ± 0.04	27.6 ± 0.2	22.0 ± 0.2
B1–28	2906 ± 36	5817 ± 11	2502 ± 8	172 ± 2	4677 ± 57	3.27 ± 0.04	6.55 ± 0.08	36.0 ± 0.4	29.3 ± 0.4
B1–29	2127 ± 33	3534 ± 9	2136 ± 4	140 ± 2	3127 ± 46	2.19 ± 0.03	4.38 ± 0.06	25.2 ± 0.4	19.4 ± 0.3
B1–30	1915 ± 16	4500 ± 7	1720 ± 3	141 ± 1	3519 ± 26	2.46 ± 0.02	4.93 ± 0.04	26.7 ± 0.2	22.1 ± 0.2
B1–31	2696 ± 64	5872 ± 15	2252 ± 7	181 ± 2	4595 ± 54	3.22 ± 0.04	6.43 ± 0.08	34.9 ± 0.4	28.8 ± 0.3
B1–32	2079 ± 35	4051 ± 9	2036 ± 5	135 ± 2	3393 ± 52	2.38 ± 0.04	4.75 ± 0.07	26.7 ± 0.4	21.2 ± 0.3
B1–33	2674 ± 62	6049 ± 15	2314 ± 6	168 ± 2	4730 ± 59	3.31 ± 0.04	6.62 ± 0.08	35.9 ± 0.4	29.6 ± 0.4
B1–34	2103 ± 30	5009 ± 9	2018 ± 4	160 ± 1	3964 ± 27	2.78 ± 0.02	5.55 ± 0.04	30.3 ± 0.2	24.8 ± 0.2

(Continued)

Table 1. Continued

Sample code	Activity concentrations A (Bq kg^{-1})*				Absorbed dose, D ($nGy \cdot h^{-1}$)	Effective dose for inside gallery	Effective dose for outside gallery	Internal Exposure Index	External Exposure Index
	^{238}U	^{232}Th	^{226}Ra	^{40}K	D (nGy h^{-1})	ED_{in} (mSv y^{-1})	ED_{out} (mSv y^{-1})	H_{in}	H_{ex}
B1–35	2077 ± 61	5034 ± 12	2029 ± 6	152 ± 3	3984 ± 80	2.79 ± 0.06	5.58 ± 0.11	30.4 ± 0.6	25.0 ± 0.5
B2–1	4228 ± 22	5463 ± 8	4580 ± 7	158 ± 1	5422 ± 36	3.80 ± 0.03	7.59 ± 0.05	45.9 ± 0.3	33.5 ± 0.2
B2–2	3225 ± 30	4797 ± 8	3047 ± 5	184 ± 1	4313 ± 26	3.02 ± 0.02	6.04 ± 0.04	35.0 ± 0.2	26.8 ± 0.2
B2–3	4420 ± 17	1543 ± 4	4531 ± 6	44 ± 1	3027 ± 36	2.12 ± 0.02	4.24 ± 0.05	30.5 ± 0.4	18.2 ± 0.2
B2–4	4399 ± 73	4406 ± 13	3804 ± 9	156 ± 3	4425 ± 87	3.10 ± 0.06	6.20 ± 0.12	37.6 ± 0.7	27.3 ± 0.5
B3–1	948 ± 35	7373 ± 12	426 ± 2	255 ± 1	4661 ± 30	3.26 ± 0.02	6.53 ± 0.04	30.8 ± 0.2	29.7 ± 0.2
B3–2	462 ± 43	2514 ± 7	319 ± 1	1057 ± 3	1710 ± 9	1.20 ± 0.01	2.39 ± 0.01	11.7 ± 0.1	10.8 ± 0.1
B3–3	655 ± 30	5240 ± 10	439 ± 2	611 ± 2	3393 ± 20	2.38 ± 0.01	4.75 ± 0.03	22.7 ± 0.1	21.5 ± 0.1
B3–4	1046 ± 41	3624 ± 9	903 ± 3	213 ± 2	2615 ± 27	1.83 ± 0.02	3.66 ± 0.04	18.9 ± 0.2	16.5 ± 0.2
B4–1	681 ± 25	1959 ± 5	616 ± 2	861 ± 3	1504 ± 8	1.05 ± 0.01	2.11 ± 0.01	11.1 ± 0.1	9.4 ± 0.1
B4–2	622 ± 90	3851 ± 7	342 ± 2	1149 ± 5	2532 ± 19	1.77 ± 0.01	3.54 ± 0.03	17.0 ± 0.1	16.0 ± 0.1
B4–3	596 ± 41	2535 ± 7	511 ± 2	1072 ± 3	1812 ± 10	1.27 ± 0.01	2.54 ± 0.01	12.8 ± 0.1	11.4 ± 0.1
B4–4	226 ± 9	647 ± 2	212 ± 1	1017 ± 2	531 ± 3	0.37 ± 0.00	0.74 ± 0.00	3.9 ± 0.0	3.3 ± 0.0
B5–1	1369 ± 58	6632 ± 16	1087 ± 7	291 ± 3	4520 ± 56	3.16 ± 0.04	6.33 ± 0.08	31.5 ± 0.4	28.6 ± 0.4
B5–2	1374 ± 38	3925 ± 10	986 ± 4	120 ± 2	2831 ± 49	1.98 ± 0.03	3.96 ± 0.07	20.5 ± 0.4	17.8 ± 0.3
B5–3	1284 ± 59	6615 ± 15	868 ± 4	191 ± 2	4404 ± 51	3.08 ± 0.04	6.17 ± 0.07	30.3 ± 0.4	27.9 ± 0.3
B5–4	1489 ± 37	6704 ± 13	964 ± 3	197 ± 1	4503 ± 28	3.15 ± 0.02	6.30 ± 0.04	31.1 ± 0.2	28.5 ± 0.2
B6–1	373 ± 16	2258 ± 4	141 ± 1	1024 ± 2	1472 ± 11	1.03 ± 0.01	2.06 ± 0.02	9.7 ± 0.1	9.3 ± 0.1
B6–2	331 ± 31	2312 ± 4	111 ± 1	1001 ± 3	1489 ± 14	1.04 ± 0.01	2.09 ± 0.02	9.7 ± 0.1	9.4 ± 0.1
B6–3	379 ± 36	2443 ± 7	156 ± 1	1045 ± 4	1591 ± 13	1.11 ± 0.01	2.23 ± 0.02	10.5 ± 0.1	10.1 ± 0.1
B6–4	401 ± 39	2307 ± 6	147 ± 2	1057 ± 4	1505 ± 22	1.05 ± 0.02	2.11 ± 0.03	9.9 ± 0.1	9.5 ± 0.1
B6–5	276 ± 30	2167 ± 5	98 ± 1	1021 ± 3	1397 ± 15	0.98 ± 0.01	1.96 ± 0.02	9.1 ± 0.1	8.8 ± 0.1
B7–1	63 ± 23	431 ± 2	62 ± 0.3	365 ± 1	304 ± 2	0.21 ± 0.00	0.43 ± 0.00	2.1 ± 0.0	1.9 ± 0.0
B7–2	66 ± 8	410 ± 1	52 ± 0.3	372 ± 1	287 ± 2	0.20 ± 0.00	0.40 ± 0.00	1.9 ± 0.0	1.8 ± 0.0
B7–3	82 ± 16	476 ± 2	65 ± 1	356 ± 1	332 ± 5	0.23 ± 0.00	0.47 ± 0.01	2.3 ± 0.0	2.1 ± 0.0
B7–4	80 ± 25	477 ± 2	64 ± 1	372 ± 1	333 ± 5	0.23 ± 0.00	0.47 ± 0.01	2.3 ± 0.0	2.1 ± 0.0

Note: Activity concentrations were given together with statistical counting uncertainties within $\pm 1\sigma$ confidence interval (68% confidence level. Total uncertainties were calculated using the law of propagation of uncertainty according to EURACEM/CTAC Guide[24].

Table 2. Range and mean values of absorbed dose rate, annual external effective doses for inside gallery and outside gallery and radiation exposure indices for the locations in Beylikova complex ore deposit in Eskişehir, Turkey.

Sample Code	D (nGy h^{-1})		ED_{in} (mSv y^{-1})		ED_{out} (mSv y^{-1})		H_{in}		H_{ex}	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean
B1.1–1.24	1498–4378	2992	1.05–3.06	2.09	2.10–6.13	4.19	12.9–37.6	25.0	9.2–27.5	18.5
B1.25-B1.35	3127–4730	3834	2.19–3.31	2.68	4.38–6.62	5.37	25.2–36.0	29.6	19.4–29.6	24.0
B2.1-B2.4	3027–5422	4297	2.12–3.80	3.01	4.24–7.59	6.02	30.5–45.9	37.2	18.2–33.5	26.5
B3.1-B3.4	1710–4661	3095	1.20–3.26	2.17	2.39–6.53	4.33	11.7–30.8	21.0	10.8–29.7	19.6
B4.1-B4.4	531–2532	1595	0.37–1.77	1.12	0.74–3.54	2.23	3.9–17.0	11.2	3.3–16.0	10.0
B5.1-B5.4	2831–4520	4065	1.98–3.16	2.85	3.96–6.33	5.69	20.5–31.5	28.4	17.8–28.6	25.7
B6.1-B6.5	1397–1591	1496	0.98–1.11	1.05	1.96–2.23	2.09	9.1–10.5	9.8	8.8–10.1	9.4
B7.1-B7.4	287–333	314	0.20–0.23	0.22	0.40–0.47	0.44	1.9–2.3	2.1	1.8–2.1	2.0

estimated from the measured activities of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in samples:

$$H_{ex} = \left[\frac{A\left(^{226}Ra\right)}{370\ Bq/kg} + \frac{A\left(^{232}Th\right)}{259\ Bq/kg} + \frac{A\left(^{40}K\right)}{4810\ Bq/kg} \right] \quad (6)$$

Normally, if the H_{ex} index is less than unity, the annual effective dose due to radioactivity in the material will be considered safe. In addition to the H_{ex} index, the inhaled radon and its short-lived progeny also pose a possible risk exposure to respiratory organs. For instance, ICRP retains the upper value of 10 mSv (effective dose, converted by convention from 600 Bq m^{-3} ^{222}Rn in dwellings for the annual dose reference level as given in ICRP 65 publication[21]). Internal exposure to radon and its progeny can also be quantified using the internal exposure index (i.e. internal health hazard index) H_{in}, from the measured activities of ^{226}Ra, ^{232}Th and ^{40}K radionuclides:

$$H_{in} = \left[\frac{A\left(^{226}Ra\right)}{185\ Bq/kg} + \frac{A\left(^{232}Th\right)}{259\ Bq/kg} + \frac{A\left(^{40}K\right)}{4810\ Bq/kg} \right] \quad (7)$$

If the exposure index H_{in} is unity, then it corresponds to 1.5 mSv/y by taking the radiation weighting factor of 1 for gamma-rays; however, if it is greater than unity, say, a factor of 10–20 times higher, the utilisation of a mine working area to be considered requires radiation protection measures for a more safe workplace.

From the measured activity results, the possible external gamma doses and health hazard indexes were calculated for the mineworkers in case opencast mining. The calculated results are given in Table 1.

The highest external exposure index is estimated to be $H_{ex} = 33.5 \pm 0.2$ (mean: 18.2 ± 0.3), and the highest internal exposure index is $H_{in} = 45.9 \pm 0.3$ (mean: 22.9 ± 0.4).

The mean absorbed gamma dose (nGy/h) inside the gallery is 52 times higher than annual world average exposure of 57 nSv/h[27]. Inside the gallery, mean $H_{ex} = 18.5 \pm 4.4$ (its median value is 19) and mean $H_{in} = 25.0 \pm 5.2$ (its median value is 25.2). The maximum annual external effective gamma dose rate was calculated to be 3.80 ± 0.03 mSv y^{-1} (mean: 2.04 ± 0.03 mSv y^{-1}) for inside gallery and 7.59 ± 0.05 mSv y^{-1} (mean: 4.08 ± 0.05 mSv y^{-1}) for outdoor (outside the e gallery. Thus, the study area can be considered as a relatively higher natural background radiation area.

For the sake of simplification, the ranges and mean values are given in Table 2 for dose rate, annual external effective doses and internal and external exposure indices for each sampling location in Beylikova complex ore deposit in Eskişehir, Turkey.

As seen in Table 2, the ore samples coded as B1–25 and B1–35 were taken from inside the gallery (in Devebağırtan sector) and pose a relatively high possibility radiation exposure level for miners because external exposure index range of $H_{ex} = 19.4$–29.6.

In the literature, the mean value for external annual effective doses received by workers in mining of rare earth ore has been reported to be 0.24–1 mSv y^{-1}[22], assuming eight working hours per day and 251 days per year. Therefore, the present result for external annual mean dose calculated for Beylikova complex ore deposit is about 2–6 times greater than the value of the literature for the case of an opencast mining.

DISCUSSION AND CONCLUSIONS

From the obtained results, the estimated doses for the mine workers are dominantly due to ^{232}Th because of high-energy gamma-rays from its decay products

and relatively higher activity content. It is also worth noting that ^{226}Ra -decay products also contribute to the estimated annual external effective doses because of a non-negligible amount of uranium in Beylikova complex ore deposit.

In this study, we measured specific activities of^{238}U, ^{226}Ra ^{232}Th and ^{40}K and thus provide a dose estimation for the mining workers. It is providing unique valuable data to the licensing holder (Eti Maden Corp.) before the mining operation starts for rare earth element ore oxides at the complex ore deposit in Beylikova-Sivrihisar site. The pilot facility in this area is already under construction.

The average effective dose limit stipulated by national radiation protection regulations for mining workers is < 20 mSv/y on an average. An effective dose limit for workers (due to occupational exposure) should not exceed 20 mSv/y over five subsequent years, and it also should not exceed the maximum effective dose 50 mSv in a single year[19,28].

From standpoint of radiation protection for mining workers, this study indicated that not only thorium, mined as a by-product, but also uranium-radium content in Beylikova complex ore poses a considerable radiation exposure level to the miners when we considered only exposed external radiation of gamma-ray emissions. It is worth noting that even if there exists a smaller amount of U-Ra in the ore, the mean external exposure index is estimated to be $H_{ex} = 18.2 \pm 0.3$, and the mean internal exposure index is $H_{in} = 22.9 \pm 0.4$. This implies that these values also should be reduced for miners even if an opencast mining process is implemented.

Some of the collected samples contain more than 0.1% wt ^{232}Th and a non-negligible amount of ^{238}U. This study gives the first results on their radioactivity contents and their possible doses to the mining workers. The present findings indicate that additional radiation protection measures need to be taken during a complex ore deposit mining process even if an opencast mining is implemented in the Beylikova-Sivrihisar site.

On the other hand, particularly radon (^{222}Rn) inhalation and even thoron (^{220}Rn) inhalation may also be a greater concern for mine workers exposed to radiation. This is another area of future work as it necessitates a continuous radon measurement for a longer period using special instruments conforming to ISO 11665–1 standard[23] as well as air filtering radioactivity measurement at the REE mine site.

The present study indicates that precautions should be taken in operational mining processes, thus mitigating occupational radiation exposure levels to miners. Occupational health problems can be reduced if special working procedures are established and personal protective equipment is used at the mining site.

Compulsory work should be done in the future for the mine galleries by using continuous mode radon measurement equipment, together with additional samples to decide radon action levels and to determine radon exhalation rates. Additionally, considering the fact that once mining operations start, other exposure scenarios will exist namely exposure to sludges, scalings and tailings,. which might have activity concentrations much higher than in the original ores and thus even higher annual dose values, these radiation problems should be considered in future works for such type of mining areas.

However, it is worth noting that the present results indicate that internal and external exposure level indices to mine workers will be lowered remarkably if an opencast REE mining process is conducted in Beylikova ore deposit by considering the necessary radiation protection measures.

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REFERENCES

1. Örgün, Y., Altınsoy, N., Şahin, S. Y., Güngör, Y., Gültekin, A. H., Karahan, G. and Karacık, Z. *Natural and anthropogenic radionuclides in rocks and beach sands from Ezine region (Çanakkale), western Anatolia, Turkey*. Appl. Radiat. Isot. **65**, 739–747 (2007).
2. Yücel, H., Solmaz, A. N., Köse, E. and Bor, D. *Methods for spectral interference corrections for direct measurements of ^{234}U and ^{230}Th in materials by gamma-ray spectrometry*. Radiat. Prot. Dosimetry **138**(3), 264–277 (2010).
3. Kaplan, H. *Eskişehir-Sivrihisar-Kızılcaören köyü yakın güneyi nadir toprak elementleri ve toryum kompleks cevher yatağı (in Turkish)*. Jeoloji Mühendisliği Dergisi, ISSN **1016–9172**(2), 29–34 (1977).
4. Öztürk, H., Hanilçi, N., Altuncu, S. and Kasapçi, C. *Rare earth element (REE) resources of Turkey: an overview of their characteristics and origin*. Bull. Min. Res. Exp. **159**, 129–143 (2019).
5. IAEA-TECDOC-1450. Thorium Fuel Cycle - Potential Benefits and Challenges, ISBN 92–0–103405–9, ISSN 1011–4289. (Vienna: IAE) (2005).

6. Gültekin, A. H., Örgün, Y. and Suner, F. *Geology, mineralogy and fluid inclusion data of the Kizilcaören fluorite–barite–REE deposit, Eskisehir, Turkey*. J. Asian Earth Sci. **25**, 629–642 (2005).
7. Yücel, H., Zümrüt, S., Narttürk, R. B. and Gedik, G. *Efficiency calibration of a coaxial HPGe detector-Marinelli beaker geometry using an ^{152}Eu source prepared in epoxy matrix and its validation by efficiency transfer method*. Nucl. Eng. Technol. **51**, 526–532 (2019).
8. DDEP, Decay Data Evaluation Project. Laboratoire National Henri Becquerel (LNHB), France, Nucleide Lara Web Database. Available on http://www.nucleide.org/Laraweb/index.php, access date: 14 November 2019
9. Yücel, H., Çetiner, M. A. and Demirel, H. *Use of the 1001 keV peak of ^{234m}Pa daughter of ^{238}U in measurement of uranium concentration by HPGe gamma-ray spectrometry*. Nucl. Instrum. Meth. Phys. Res. A **413**, 74–82 (1998).
10. Huy, N. Q. and Luyen, T. V. *A method to determine ^{238}U activity in enviromental soil samples by using 63.3 keV photopeak-gamma HPGe spectrometer*. Appl. Radiat. Isot. **61**(6), 1419–1424 (2004).
11. Yücel, H., Solmaz, A. N., Köse, E. and Bor, D. *Spectral interference corrections for the measurement of ^{238}U in materials rich in thorium by a high resolution gamma-ray spectrometry*. Appl. Radiat. Isot. **67**(11), 2049–2056 (2009).
12. Yücel, H., Köse, E., Esen, A. N. and Bor, D. *Correction methodology for the spectral interfering γ-rays overlapping to the analytical peaks used in the analysis of ^{232}Th*. Appl. Radiat. Isot. **69**, 890–897 (2011).
13. Lavi, N., Groppi, F. and Alfassi, Z. B. *On the measurement of ^{40}K in natural and synthetic materials by the method of high-resolution gamma-ray spectrometry*. Radiat. Meas. **38**(2), 139–143 (2004).
14. Sima, O., Arnold, D. and Dovlete, C. *GESPECOR: a versatile tool in gamma-ray spectrometry*. J. Radioanal. Nucl. Chem. **248**(2), 359–364 (2001).
15. SDD Detector Specifications (USA, Amptek Inc). Available on https://www.amptek.com/products/sdd-x-ray-detectors-for-xrf-eds/fast-sdd-silicon-drift-detector#Specifications access date: 20 December 2019
16. Akduman, O. *Application of fundamental Paramaters method in EDXRF Spectrocopy to the quantitative analysis of some powder and metallic samples, master thesis, Ankara University*. Institute of Nuclear Sciences pp. 1–84 (2019).
17. Akkaya, G., Yücel, H. and Budak, M. G. *Determination of manganese content in some commercial manganese ores in Turkey k0-NAA standardization method using ^{241}Am–be isotopic neutron source*. AIP Conference Proceedings **2075**, 070005 (2019). doi: 10.1063/1.5091196.
18. UNSCEAR (2000). United Nations Scientific Committee on the effects of atomic radiation. Sources and Effects of Ionizing Radiation, Vol. I (Sources). Available on https://www.unscear.org/docs/publications/2000/UNSCEAR_2000_Report_Vol.I.pdf, access date: 17 November 2019 UNITED NATIONS PUBLICATION,ISBN 92-1-142238-8, UN, New York.
19. Valentin, J., Ed. The 2007 Recommendations of the International Commission on Radiological Protection, The International Commission on Radiological Protection (ICRP) Annals of the ICRP Publication 103. Published for the International Comission on Radiological Protection by Elsevier (2007).
20. Beretka, J. and Mathew, P. J. *Natural radioactivity of Australian building materials, industrial wastes and by-products*. Health Phys. **48**, 87–95 (1985).
21. Protection Against Radon-222 at Home and at Work. *ICRP publication 65*. Ann. ICRP **23**(2), 1–48 (1993).
22. Qifan, W., Hua, L., Chenghui, M., Shunping, Z., Xinhua, Z., Shengqing, X. and Hongyan, W. The use and management of NORM residues in processing Bayan Obo ores in China. In: International Atomic Energy Agency(IAEA) Naturally Occurring Radioactive Material (NORM VI) Proceedings of an International Symposium Held in Marrakech. (Morocco), 22–26 March 2010, STI/PUB/1497, 978-92-0-113910-8) pp. 65–78 (2011).
23. ISO 11665-1. Measurement of Radioactivity in the Environment - Air: Radon-222- Part 1: Origins of Radon and its Short-Lived Decay Products and Associated Measurement Methods. International Organization for Standardization(ISO), CEN-CENELEC Management Center: Rue de la Science 23, B-1040 Brussels (2019).
24. EURACEM/CITAC Guide CG 4. In: Quantifying Uncertainty in Analytical Measurement, 2nd edn. Ellison, S. L. R., Rosslein, M. and Williams, A., Eds. Eurachem/CITAC Working Group, Published by Eurachem (2000) ISBN 0 948926 15 5.
25. UNSCEAR, 2008. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and Effects of Ionizing Radiation, Vol. I. (Sources). Available on https://www.unscear.org/docs/publications/2000/UNSCEAR_2000_Report_Vol.I.pdf, access date: 17 November 2019. UNITED NATIONS PUBLICATION, ISBN 978 9211422740, UN, New York.
26. Rao, D. D. *Use of hazard index parameters for assessment of radioactivity in soil: a view for change*. Radiat Protection Environ **41**, 59–60 (2018).
27. Dragovic, S., Jankovic, L. J. and Onjia, A. *Assessment of gamma dose rates from terrestrial exposure in Serbia and Montenegro*. Radiat. Prot. Dosimetry **121**, 297–302 (2006).
28. IAEA Safety Standards Radiation Protection and Safety of Radiation Sources. International Basic Safety Standards, General Safety Requirements Part 3, No. GSR Part 3. (Vienna: IAEA) (2011).